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RESEARCH AND DEVELOPMENT
TECHNICAL REPORT ECOM-0287-F

METALLIZING AND EVALUATION OF BORON NITRIDE MICROWAVE WINDOWS

FINAL REPORT

by

Oskar Heil

April 1969

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UNITED STATES ARMY ELECTRONICS COMMAND • FORT MONMOUTH, N.J. 07703

Contract DAAB07-67-C-0287
HEIL SCIENTIFIC LABORATORIES, INC.
Belmont, California

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METALLIZING AND EVALUATION OF BORON NITRIDE MICROWAVE WINDOWS

Final Report

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U.S. Army Electronics Command
Fort Monmouth, New Jersey

DISTRIBUTION STATEMENT

ABSTRACT

The objectives of this program are the application of the metallizing and bonding techniques developed by Heil Scientific Laboratories under USAECOM Contract DA28-043-AMC-00429(E)[1] to the fabrication and evaluation of high power S-band microwave window assemblies using chemical vapor deposited isotropic boron nitride (CVD-BN) as the window dielectric. Four S-band isotropic CVD-BN pokerchip microwave windows have been furnished by the Techniques Branch, Electron Tubes Division, USAECOM to the Heil Scientific Laboratories for this program.

Isotropic boron nitride is the best dielectric for high power microwave windows because of its low dielectric constant, low dielectric losses, high dielectric strength, good heat conductivity and lack of multipactoring. It, however, is difficult to metalize on account of its low thermal expansion, its relative low mechanical strength and its different chemical behavior as a nitride compared with the usual window materials, which are oxides.

Aluminum shows excellent wetting of boron nitride at 1100°C in vacuum. BN can be brazed to itself and to graphite at this temperature.

After particle bombardment coating with aluminum, these brazes can be made at 700°C with good adherence.

Vacuum pressure seals to aluminum made at 500 to 600°C with or without prior metallizing are strong and vacuum tight.

Aluminasilicate glasses like Corning 1720 can also be vacuum pressure sealed to aluminum and copper at 600°C on account of their high softening temperature. These glasses match the thermal expansion of isotropic BN. Aluminum pressure seals have been made to boron nitride with aluminasilicate glass as backing.

Aluminasilicate glass of the proper dimensional size could not be obtained from commercial sources expeditiously and economically; therefore, a substitute glass was chosen. Pyrex glass was tested and good pressure seals to copper and aluminum were made. Aluminum pressure seals to boron nitride with Pyrex backing are also successful inspite of a small difference in thermal expansion.

On Pyrex windows of 3.5 inch diameter the aluminizing of windows and Pyrex backing rings as well as the pressure sealing of the complete window assembly with copper and aluminum were practiced and perfected, resulting in vacuum tight strongly bonded units.

Unfortunately, a test of a complete CVD-BN window assembly using the techniques described could not be achieved within the time and financial resources of this contract, though it is believed the work accomplished here will be of value in future CVD-BN microwave window programs.

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1.0 FACTUAL DATA

1.1 ISOTROPIC BORON NITRIDE AS MICROWAVE WINDOW MATERIAL

At the Raytheon Research Laboratory, Waltham, Mass., S. R. Steele, J. Pappis, R. Ellis and L. Hagen have developed a new insulating material, isotropic pyrolytic boron nitride. This material is definitely an excellent dielectric for high power microwave windows; for high average power as well as for high pulse power. This becomes evident by studying Table V, "Properties of Some Insulating Materials" of Technical Report ECOM-01343-4[2] July 1966, Pages 22 and 23 where the physical data of isotropic CVD BN commercial BeO, Al_2O_3 anisotropic CVD BN and commercial hot pressed BN are compared. The window problems are thoroughly discussed on Pages 19 to 21 of the same report and on Pages 8 and 9 of the Technical Report ECOM-01343-5[3] October 1966. The only thing which should be added is the experimental experience which R. Bierce had in checking an anisotropic BN window under pulsed operation in the Ring Resonator of the Stanford Linear Accelerator. This experiment was done several years ago and showed at low power level a weak multipactor which disappeared at higher power level. This window had no multipactor suppression coating. More recent results obtained by

R. Bierce show multipactor suppression on alumina windows by coating them with boron nitride. This adds another good property to those shown in Table V of the Raytheon report. A few of these values given for isotropic CVD BN shall be mentioned: Dielectric Constant 3.36-3.01, Loss Tangent 0.6×10^{-4} at room temperature diminishing to 0.5×10^{-4} at $1000^{o}C$, Electric Resistivity $10^{15}\,\Omega\,cm$ which is an order of magnitude better than other materials, Thermal Conductivity of 0.045 $cal/cm^2/sec/^{o}C$ a low Thermal Expansion of $4 \times 10^{-6}/^{o}C$. The only property where this material is inferior to others is the mechanical strength with 14,000-17,000 psi. This relative low strength combined with the small thermal expansion makes the sealing of high frequency windows a problem. In addition, the chemical affinity of a nitride is quite different from the normally used oxide window materials.

1.2 METAL SEALING OF ISOTROPIC CVD BORON NITRIDE WINDOWS

It is the task of this research project to find a suitable solution for a low loss, mechanically strong metal seal to this excellent window material. The new sealing method shall be tested on high pulse power S-band windows at the facilities of the Stanford Linear Accelerator.

First the mechanical material properties involved in the seal shall be discussed. Boron nitride has the same atomic structure as graphite with C-atoms being alternately replaced by B- and N-atoms. In the Siemens Laboratories some thirty years ago a systematic study was made on the hardness of high purity graphite in dependence of its crystalite size, with the results that the hardness of carborundum could be reached on very fine crystaline graphite. This work was published in the "Siemens Berichte". The results are easily understood if we are aware that the carbon to carbon bond in graphite is stronger than in diamond. The C-C distance in the hexagonal plane of graphite is 1.42 Å against 1.54 Å for diamond. This stronger bond in graphite is also evident from the heat conductivity of pyrolytic graphite in the direction of the hexagonal planes, which is 1.0 cal/cm^2/sec/$^{\circ}$C against the conductivity in diamond of 0.8 cal/cm^2/sec/$^{\circ}$C.

Another evidence of the strong bond in the layer structure of pyrolytic graphite and anisotropic CVD BN is the negative thermal expansion in this direction for temperatures below 150°C for both substances. (See Figure 4, Page 18 of Technical Report ECOM-01343-1).[4] These layer type materials share this property with fused silica, a fiber type

material with the $Si-O_2-Si$ fibers showing negative thermal expansion below $-70^{\circ}C$. The more violent heat vibrations perpendicular to these fibers or planes causes them to shrink in the direction of their strong bonds, which generates the negative thermal expansion. If this vibration, however, become too violent at higher temperatures, they start interfering with the neighbor planes. Those planes get more and more interhooked by the vibrations. The free gliding of planes gets inhibited, which strengthens the material. Graphite and boron nitride show in contrast to other solids a strengthening or at least no weakening with increasing temperature. This is due to that interhooking of the planes, which in themselves are stronger than diamond. It also explains that these peculiar materials can become very hard if only the crystallites are small and oriented at random. Crystal plane slippage in one crystal is stopped by its neighbor, explaining the hardness of carborundum in graphite and the increased flexural strength in isotropic CVD boron nitride. The perfect isotropy of boron nitride produced from organic boron compounds is demonstrated in Figure 8, Page 27 of Technical Report ECOM-01343-1[4] and the small, uniform crystallite size of one-quarter micron

is shown in Figure 6, Page 23 of the same report. Without the increase in tensile strength resulting from the isotropy and the fine crystaline structure of this boron nitride a window metal seal would not be practical. Another property of the material helps making a seal easy. The Young's Modulus of BN is only 2.5×10^6 psi. The great elastic softness reduces possible stresses which develop if a material is sealed to it which does not perfectly match the thermal expansion.

1.3 MATERIALS MATCHING THE THERMAL EXPANSION OF ISOTROPIC CVD BORON NITRIDE

Of the metals only tungsten matches the thermal expansion of isotropic CVD BN closely. This material has, however, other disadvantages. The elastic modulus is with 57.5×10^6 psi, twenty-three times that of isotropic CVD BN. Tungsten is hard and brittle and difficult to manufacture in the desired ring form suitable for microwave windows.

Other materials matching boron nitride are the different tungsten sealing glasses. A few of them are shown in the following Table with the thermal expansion co-efficient in $10^{-7}/^{\circ}C$, Strain Annealing and Softening Points in $^{\circ}C$ and Modulus of Elasticity $\times 10^6$ psi. The latter is approximately

4.5 times lower than that of tungsten. Besides low elasticity and thermal expansion match to BN ($40 \times 10^{-7}/^{o}C$) we are interested in high softening temperatures, which allow metal sandwich pressure seals to be made at temperatures around $600^{o}C$. The glasses are listed in the Table according to their softening temperatures.

TABLE OF POSSIBLE GLASSES FOR PRESSURE SEALS

Corning No.		Thermal Expansion $10^{-7}/^{o}C$	Strain Point ^{o}C	Anneal. Point ^{o}C	Softening Point ^{o}C	Modulus of Elasticity 10^{6} x psi
1715	Alumino Silicate	35	834	866	1016	12.4
1720	"	42	-	715	-	-
1710	"	42	672	712	915	13.1
1723	"	46	670	708	910	12.8
7991	Tungsten Sealing	41	525	565	800	-
7740	Pyrex	32	515	555	820	10.1
9700	UV Glass Corex	37	512	558	804	-
3320	Uranium	40	497	555	780	-

As can be seen from the Table, the first four glasses which are all aluminosilicate glasses are most favorable for seal backing rings. 1720 and 1723 were used successfully in aluminum pressure seals with CVD BN. The other alumino-silicate glasses were not available but should work just as good, whereas the remaining glasses must be sealed at lower temperatures to prevent

deformation. Pyrex to CVD BN seals are successfully made at 505°C and are used on the actual window seals.

1.4 ALUMINUM ON GRAPHITE

Three years ago we made an interesting observation when vaporizing aluminum from graphite. It is known that aluminum forms with graphite the aluminum carbide $Al_4 C_3$ which is hygroscopic and decays in air forming methane. We wanted to see how the carbide formation competes with aluminum vaporization in vacuum. A flat piece of graphite on which a small piece of aluminum was placed was high-frequency heated under a vacuum bell jar. The aluminum did melt forming a non wetting drop. When the temperature of 1100°C was reached the aluminum started wetting the carbon, spreading in a thin film over the carbon surface. At the same time aluminum started vaporizing appreciably. The temperature was lowered immediately. The bond between aluminum and carbon was very strong. Assuming that $Al_4 C_3$ formation was responsible for the bond, the piece was submerged in water for several days. But there was no deterioration or decomposition taking place and the piece survived three years in air with no change. Graphite was brazed to graphite at 1100°C and a composite material of granular

graphite with aluminum was produced which was also stable. At higher temperatures the expected $Al_4 C_3$ formation takes place. Yellow crystals of this material can be seen. The remaining graphite deteriorates into dust in air.

From these observations we drew the conclusion that another lower aluminum carbide must exist which is stable against moisture and which adheres strongly to graphite and aluminum. A search in literature showed that no such compound is known.

1.5 ALUMINUM ON ISOTROPIC CVD BORON NITRIDE

In view of the great structural and chemical similarity between graphite and boron nitride the same experiments described in 1.4 were carried out with CVD BN and aluminum with the same results as obtained on graphite. A flat piece of boron nitride was heated inside of a deep graphite crucible. To prevent rolling off of the aluminum drop, a small hole was drilled in the boron nitride to retain the liquid aluminum. The aluminum remained non wetting as a drop in this hole until 1100°C. Then it started spreading over the top surface, down the side surfaces reaching the bottom surface in about ten seconds, and then the temperature was reduced to prevent further spreading. The piece is shown on Figure 1 in top and bottom views. On the corners where the CVD BN touched the graphite, aluminum spread over to the crucible and brazed the BN to the

graphite from which it was broken loose. (See left corner on lower picture of Figure 1). On the edge of the spreading aluminum film a grey discoloration of the boron nitride indicates the bonding reaction product of the two materials. We do not know if this interface material is conducting or insulating, but we can assume with certainty that it will be a more lossy conductor than aluminum and a more lossy dielectric than boron nitride. We are, therefore, interested in keeping its thickness at a low value. In the experiment described, the reaction was produced thermally. It has been shown[1] that particle bombardment bonding has practically no penetration depth and still maintains great bonding strength. From the thermal experiment we know that good bonding exists. Therefore, we turned to particle bombardment bonding of aluminum to isotropic CVD boron nitride.

1.6 PARTICLE BOMBARDMENT BONDED ALUMINUM ON BORON NITRIDE

Isotropic CVD Boron nitride flat pieces were aluminized using the standard procedure as described in the Technical Report ECOM-00429-F[1] on Page 18. Off sputtering for surface cleaning was run for 5 minutes with a self generated d.c. voltage of 200 Volt on the sample holder. Aluminum coating followed for 15 minutes with 240 Volt on the aluminum target. On

this prepared aluminized surface vacuum molten aluminum adheres without the use of high temperature. Figure 2 illustrates a drop of molten aluminum heated to 700°C. The sample is cut apart through the middle of the drop. Despite the great thickness of aluminum and the great difference in thermal contraction the bond did not break. Similar experiments with graphite and aluminum show the same result.

Figure 3 shows in a) two graphite and in b) two boron nitride pieces, sputter coated and brazed with aluminum. After vacuum leak checking, which showed the samples to be vacuum tight, they were cross-sectioned as shown in Figure 3.

1.7 PRESSURE SEALING OF CVD BORON NITRIDE AND ALUMINOSILICATE GLASS TO ALUMINUM

It was assumed that the high temperature of 1100°C necessary for aluminum to wet boron nitride or graphite is only needed for breaking the oxide skin on the aluminum. In a pressure seal the flowing of the metal breaks the oxide skin, replacing the effect of the high temperature.

Pressure seals made of aluminum to unmetallized isotropic CVD boron nitride or aluminosilicate glass and graphite at 600°C were all vacuum tight and mechanically strong. Figures 4 and 5 show sandwich seals of isotropic CVD BN to Corning glass 1723 with

aluminum of .010" original thickness in between. In Figure 4 the boron nitride is broken away with force. The remaining fragments of CVD BN on the bond surface of the aluminum indicate the bonding strength. Cross-sectioning reveals the aluminum deformation. The specific pressure in these seals was 300 kg/cm^2 the pressing time 60 minutes, the temperature 600°C. Figure 6 shows a CVD BN to CVD BN pressure seal, which was vacuum tight and then torn apart to illustrate the good bonding strength between aluminum and the boron nitride, which in this case was anisotropic. The pealed off BN is clinging to the aluminum foil. Figure 7 illustrates good bonding of an aluminosilicate glass pressure seal with aluminum which was intentionally broken. The glass adheres strongly to the aluminum and one crack in front goes straight through both glass rings indicating that the seal was free of stress. The high purity aluminum (99.998%) used in these tests has an annealing temperature of 20°C. Figure 8 shows one of the many successful glass-copper-glass pressure seals which were made at the same temperature of 600°C with 60 minutes pressing at 300 kg/cm^2. The copper was 0.006" thick. Copper-glass seals are necessary for the vacuum tight aluminum-copper transition in the final window design (Figure 10). It should be mentioned that the glass remains rigid and

only the copper deforms. Glass to glass surfaces do not seal at this temperature and pressure.

1.8 HYDRAULIC PRESSING IN HIGH-VACUUM AT ELEVATED TEMPERATURES

The pressure seals mentioned were made in the vacuum press, illustrated in Figure 9. A 500 liter/sec mercury diffusion pump with a freon 22 cold trap produced the vacuum. The pressure cylinder is housed in the bottom part of the water cooled metal vacuum container. The pressure is hand pumped. In the upper part is the pressure region with a tantalum tube vacuum furnace, the temperature of which can be measured with an optical pyrometer through the quartz window. The glass top which can be removed for loading contains a heat shield and a liquid nitrogen trap. This trap was found to be unnecessary. The mercury pressure was sufficiently reduced by the freon trap.

1.9 THE BORON NITRIDE WINDOW CHAMBER

The boron nitride window chamber as it is planned for testing in the Stanford Linear Accelerator test ring will be of the type shown in Figure 10. The isotropic CVD boron nitride poker-chip window will be aluminized on its edge, that is the cylindrical surface and the flat surfaces as far as they will be

covered by the aluminized aluminosilicate glass rings. These glass rings will be aluminized on the surfaces where they touch the window and on their inner cylindrical surfaces. The surfaces touching the copper L-rings are bare as are the two outer glass rings which serve only as backing rings to the copper seals. The high frequency field will no where reach the glass. The high frequency current flows over the copper L-rings, over the aluminum on the inner surface of the glass rings and on the outer surface of the window. The combination of copper L-rings, glass rings and window will be sealed in one vacuum pressure operation. This combination will be inserted into the window chamber. Only the good conducting metals, copper and aluminum will be used with practically no interface between aluminum and the isotropic CVD boron nitride.

1.10 BACKING RING MATERIALS

In the just discussed window geometry of Figure 10, no high frequency field reaches the backing ring material and no high frequency current flows in the metal dielectric bonding area of the backing ring material. No dielectric or ohmic losses must be considered as long as the conductive coatings (aluminum and copper) are thicker than the high frequency current penetration depth (skin effect). The only properties we must consider in selecting the backing

ring material are: 1) thermal expansion 2) elastic modulus, 3) tensile strength, 4) softening temperature for pressure sealing. The most favorable material would be some Zircon-silicate ceramic with a suitable thermal expansion, like Alsimag 475, American Leva Corp. (43.10^{-7}) or Berliner Hartporcellan, Staatliche Porzellanmanufaktur Berlin ($38.10^{-7}/C^{o}$). These rings can be metalized with the usual molybenum manganese process and brazed to the copper rings before aluminizing and pressure sealing to the boron-nitride window. The copper pressure seal is eliminated and replaced by a brazed seal. The next best materials are the alumina silicate glasses, Corning 1720 or 1710 requiring pressure seals for copper and aluminum which are done in the same operation. The pressure temperature can be between 600^{o}C and 650^{o}C without deforming the glass.

Both materials, the ceramics and the glasses are not readily available in the right size or in small quantities. We are therefor forced to use the less favorable but available Pyrex glass with a thermal expansion of 32×10^{-7} C^{o}. The highest allowable pressing temperature on this softer glass was found to be 505^{o}C. Strong and vacuum tight seals to copper and aluminum at pressures of 800 kg/cm^2 were obtained.

1.11 PYREX GLASS PRESSURE SEALING TO ALUMINUM AND COPPER

The hydraulic vacuum pressure arrangement shown in Figure 9 was used to find the proper temperatures and pressures for sealing pyrex glass to copper and aluminum. Seals similar to those shown in Figures 4, 5, 7 and 8 were made. The temperature was measured with a copper-constantan thermocouple attached to the copper sealing disks. A temperature of 505°C could be used with pressures of 800 kg/cm^2 for one hour with glass deformation of about 4 per mille of its length. The copper is oxidized in nitrogen with a few percent oxygen at 900°C. The thickness of copper is .006" and of aluminum .001" which gives good seals for both metals under the same temperature and pressure conditions.

1.12 WINDOW AND BACKING RING ALUMINIZING

The window edge and the first backing ring need an aluminizing which is thick enough to conduct all the high frequency current, without penetration of magnetic fields to the outside. The half-value thickness for 3000 MHz is 0.26 milligrams/cm^2 for aluminum. The coating thickness should not be less than 1 milligram/cm^2. The vapor deposition was done while the window or backing ring were rotating around a vertical axis. Figure 13 shows the heating and

masking arrangements for the window on the left and for the backing ring on the right. The window is sandwiched between two graphite disks. Gliding of the disks and window against each other is prevented by thin tungsten wire hooks touching the rim of the window and fastened with screws to the graphite. The unit is high frequency heated and the aluminum vapor is deposited on the window at about 600^{o}C from three vapor sources spaced 3 to 4 mm slightly below the window rim, coating the lower flat and the cylindrical surface. The window was then turned upside down and coated again. In the same way the backing ring is coated twice. It is held inside a graphite ring resting on a small step at the bottom end. The graphite ring is held by sapphire rods on the rotating tantalum cylinder. The vapor sources are mounted close to the inner edge of the glass ring.

1.13 PREPARATION OF GLASS RINGS

The glass rings are sliced off a tubing of proper dimensions and surface ground to the right thickness. Glass tubings of this size are not perfectly round. The rings however deform elastically enough to slide over a close fitting, round graphite core, which is induction heated at 800^{o}C in air. Ten rings piled on top of each other are treated together

and come out perfectly round. They are then beveled on all four edges.

The vapor coating leaves the outer cylinder surface and the outer beveled surfaces clear. The aluminum coating on one of the flat surfaces is removed partially for the copper-glass seal leaving a half millimeter wide aluminum coating on the inside. This aluminum seals to the copper assuring a low loss ohmic transition for the high frequency currents. The aluminum is removed by grinding the backing ring in a stepped copper ring. The bare glass makes the vacuum seal to the copper L-ring.

1.14 COPPER AND ALUMINUM L-RINGS

The copper L-rings with a 15 mm wide cylindrical section and a 3.5 mm flat surface were made from .005" OFHC copper sheet using hydroforming.

Between the aluminized window and pyrex backing ring a cushioning aluminum ring of .001" thickness is used for the pressure seal. To keep this thin ring in shape and position, it is made in L-form with the cylindrical part fitting on the window periphery. The easiest way to make these delicate rings was found by cutting the aluminum foil on a glass surface with a roller type glass cutter (tungsten carbide) and spinning the edge into the L-shape over the rim of a window.

1.15 PRESSURE SEALING OF PYREX GLASS WINDOWS

In order to save the precious boron nitride windows, the pressure sealing of the whole window assembly with aluminum and copper seals was practiced on pyrex glass windows. These glass windows have the additional advantage that stresses within the window material can easily be observed with polarized light. Eight such windows were sealed to determine optimum temperature, pressure and time. Modifications in the pressing and heating were needed to achieve uniformity in temperature and pressure. The vacuum hot pressing arrangement for the windows is shown in Figure 11. It is shown sitting on a board on top of the 24 inch diameter stainless steel vacuum container into which it is lowered for pressing. A thin metal tubing feeds through the center of the lid the compression gas to the stainless steel bellows, which produce the pressure. Nitrogen from a cylinder was used and the pressure was admitted slowly through a capillary to a maximum pressure of 74 lb/in^2 to which 14.7 lb/in^2 from the vacuum must be added resulting in a total pressure of 88.7 lb/in^2. The mean diameter of the bellows is 8.9" giving a total pressing force of 5590 lb or 2530 kg. The sealing surface, which is the flat surface of the backing rings, has a mean diameter of 86 mm; a width of 2.5 mm

and an area of 6.75 cm^2. The pressure per square centimeter is 829 kg. The frame is made of stainless steel. Two safety tubes which can slide sideways on the t p bar for easier assembly limit the possible expansion of the bellows in case of breakage. This happened only once in the graphite rings due to one-sided pressure, before the pressure equalizing pad was introduced (Figure 12). The pressure transmitting column consists of the pressure equalizing pad on top, two kovar rings with holes for pumping and admitting the leads for the heating plates, a thermocouple; plus two Vycor cylinders, and two graphite guiding rings. In the center the window assembly as shown in finished form (see Figure 14) consisting of four pyrex backing rings, the two inner ones aluminized, the copper and aluminum L-rings and the aluminized window. The copper-constantan thermocouple is fastened with a clamp screwed on the upper copper ring. The main cylindrical heater is made of a molybednum ribbon coil, which is tied to six alumina rods and surrounded by a radiation shield. It stands with three legs and centering feet on the lower press platform but is positioned in the Figure to the front for better viewing. The hook with wire rope on top allows easy lifting and lowering of the adjusted and aligned assembly.

Shifting is made impossible: the bellows are before assembling evacuated and the atmospheric pressure is let in afterwards. The elasticity of the bellows exerts a small pressure holding everything in place.

1.16 PRESSURE EQUALIZING

It was first thought that the top plate of the bellows cushioned by the compressed gas would equalize any lateral pressure forces. From the difference in metal deformation of the L-rings, we learned that this was not so. Cross-sections like the one shown in Figure 14 showed the difference. Under this one sided pressure which is a result of stiffness of the bellows under pressure, one of the graphite rings was crushed, breaking the window and one Vycor cylinder. A flat ball bearing pad was constructed as shown in Figure 12. It consists of two hardened steel plates, an aluminum plate with hexagonal aperture as ball guide, 37 balls and six turnable flaps to arrest the gliding motion during assembly. The flaps are turned horizontal as shown in Figure 11 before pressure is applied. The new graphite rings were made of a dense and fine grained graphite (ATJ graphite, Union Carbide). Pressing with the ball bearing pad is very uniform.

1.15 TEMPERATURE EQUALIZING

Initially only the cylindrical molybdenum ribbon heater was used. The pyrex windows showed in polarized light severe stresses which lead to fracture. The central part of the window was not heated enough. Multiple copper disk heat shields on top and bottom of the window brought some improvement. Only the introduction of the two flat heating elements shown in Figures 11 with the proper balancing between cylindrical and flat heaters solved the problem, resulting in stress-free windows. Heat balancing will be less critical on boron nitride windows on account of the better heat conductivity.

1.16 ALUMINUM VAPOR DEPOSITION

The low vapor pressure of aluminum, requiring high vaporizing temperatures of 1100° to $1200^{\circ}C$ in combination with the great chemical re-activity of the metal make the deposition of coatings of the thickness we require (one milligram per square centimeter) difficult. If impurities vaporize the ductility of the deposited metal suffers.

Lucalox, a high purity alumina shows no reaction with aluminum at the vaporizing temperature. We used Lucalox tubings closed off on one end with a sapphire rod heated with a wound on tungsten spiral. Better thermal contact between heater and Lucalox was

obtained by sintering alumina heater coating on and between the turns. At the vaporizing temperature, aluminum starts wetting the Lucalox, creeping over the whole surface and spoiling the tungsten filament by alloying. This vaporizer was therefor abandoned.

The vaporizers used on the pyrex windows and rings were commercially available conical tungsten spirals, with alumina isolation (Sylvania A1236A), 7 turns, 4 mm diameter, 7 mm high, loaded with a conical core of aluminum of 80 milligrams. About 3/4 of the load was vaporized. Three coils were used to coat one side of the window or backing ring. The coils were used only once. It is difficult to prevent alumina vaporization which reduces the ductility of the deposited coating. The temperature of the heater runs too high, especially towards the end of vaporization resulting in coatings which are sometimes quite hard. We did not want to go with this rather uncontrolled coatings onto the boron nitride windows, even though they gave strong vacuum tight seals on the pyrex windows.

1.17 GRAPHITE ALUMINUM VAPORIZER

An improved vaporizer using high purity dense graphite (ATJ Union Carbide) as crucible and

carbon contact heating was developed as illustrated in Figure 15. As mentioned earlier, aluminum starts wetting graphite at 1100°C. Normal graphite being porous gets penetrated by aluminum and structurely destroyed by one vaporization. ATJ graphite is fine grained and dense showing practically no reaction with aluminum, remaining good after many evaporations. The reduced reaction might also be a result of the higher purity of this graphite.

A carbon-carbon contact does not reduce its resistance by sintering and fritting at the temperatures of 1100° - 1200°C, because carbon does not melt and recrystallation is low at this temperature. The heating effect on the contact resistance is therefor constant and dependable. The graphite vaporizer of Figure 15 is easy to build. It consists of rotational symmetric parts, which are with exception of the ATJ graphite crucible, all split into two and hinched with a turning point around the ceramic rod. The two jaws holding the crucible are ordinary graphite and are clamped with screws between copper parts which act also as radiation shields. The cross-section is drawn in scale 2:1. The power consumption while vaporizing is 350 Watt at about 3.5 Volt

and 100 Ampere. The spring in the photograph consists of tungsten wires which are isolated to prevent closed loop heating in the high frequency field, used for window and backing ring heating. (The grooves in the ceramic half shells have no significance. A coil body was used to make these parts). The graphite vaporizer produces the cleanest and most ductile aluminum deposits and would be used on the boron-nitride windows in case the work could be continued.

1.18 THE SURFACE OF BORON NITRIDE

There still remains the problem to find the best surface treatment of the windows, which is most favorable for the aluminum bonding. Air firing would produce a thin oxide layer on the boron nitride surface, whereas high temperature vacuum heating without exposure to the air before coating with aluminum would give a clean boron nitride surface which is also free of any possible water absorption. It could be best if this study of different surface treatment could precede the forming of the actual four 3.5" diameter boron nitride windows, which have been delivered to us

Metalizing of boron nitride has been restricted to the good conducting and good adhering

aluminum which has the disadvantage of requiring a light metal-heavy metal transition. This transition could become quite simple with the use of a zirconium-silicate ceramic for backing rings, which can be joined to copper by brazing before the aluminum pressure seals to the boron nitride window are made.

2.0 CONCLUSIONS

It has been found that vacuum pressure seals of aluminum to isotropic CVD boron nitride are vacuum tight and show bonding strength greater than the strength of isotropic CVD boron nitride.

Aluminosolicate glasses match the thermal expansion of isotropic CVD BN and can be pressure sealed to aluminum and copper. Pyrex glass with a less perfect expansion match to boron nitride can also be pressure sealed at a lower temperature to aluminum and copper.

The combination of the good conducting metals copper and aluminum with the most excellent dielectric material boron nitride should make the lowest loss high frequency microwave window. The sealing technique was developed and tested successfully with pyrex glass substituting the boron nitride. Lack of time and funds did not allow us to apply this technique to the isotropic boron nitride windows.

3.0 LITERATURE CITED

(1) "Particle Bombardment Bonding and Welding Investigation" - Technical Report ECOM-00429-F, by Oskar Heil, Heil Scientific Laboratories, Inc., Contract DA 28-043-AMC-00429(E), AD-658685.

(2) "Chemical Vapor Deposited Materials For Electron Tubes" - Technical Report ECOM-01343-4, by S. R. Steele, J. Pappis, L. Hagen, H. Schilling; July 1966, Raytheon Company, Contract DA 28-043-AMC-02343(E).

(3) "Chemical Vapor Deposited Materials For Electron Tubes" - Technical Report ECOM-02343-5, by S. R. Steele, J. Pappis, L. Hagen, H. Schilling; October 1966, Raytheon Company, Contract DA 28-043-AMC-01343(E).

(4) "Chemical Vapor Deposited Materials For Electron Tubes" - Technical Report ECOM-01343-1, by S. R. Steele, J. Pappis, R. Ellis, L. Hagen; October 1965, Raytheon Company, Contract DA 28-043-AMC-01343(E).

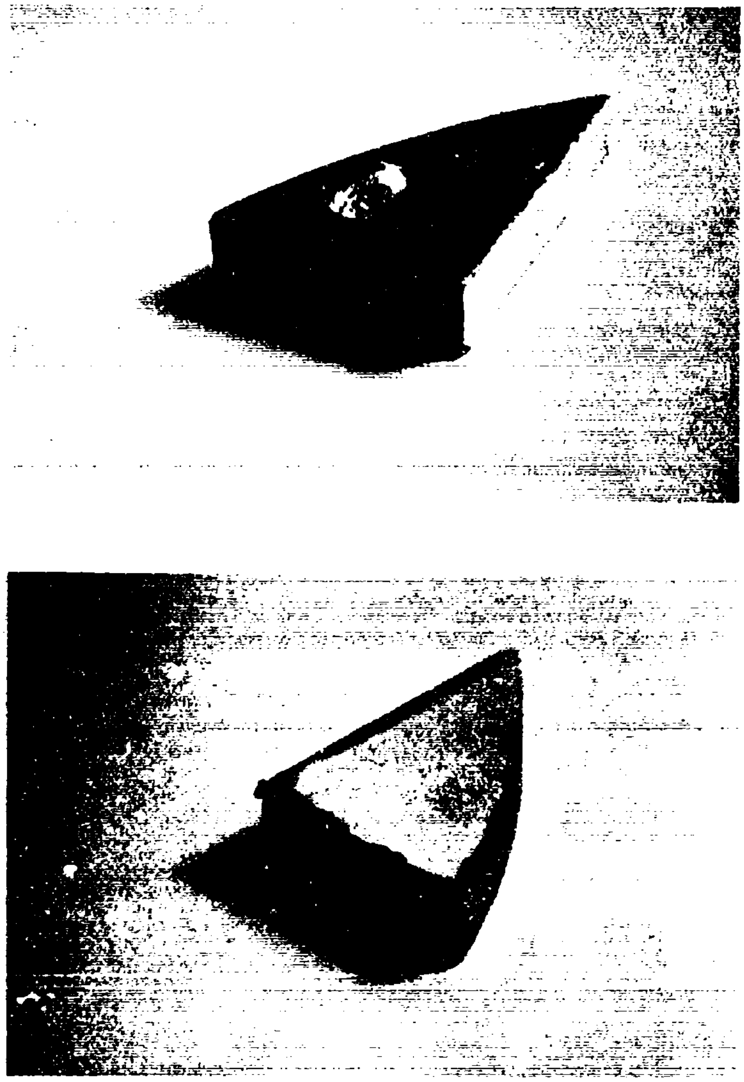


FIGURE 1

CVD Boron-Nitride Wetted by Aluminum at 1100°C

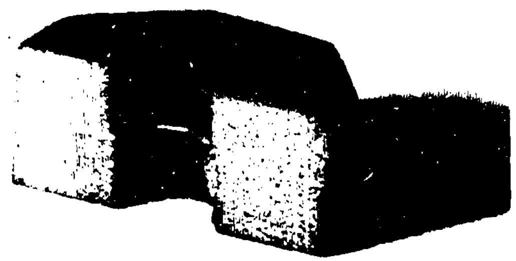
Aluminum molten in hole (top picture) spreads as thin film over top and side surfaces, reaching the bottom surface (lower picture) in about 10 seconds. Spreading is stopped by lowering the temperature.

FIGURE 2

Isotropic CVD Boron-Nitride Sputter-Coated with Aluminum. Drop of Aluminum Vacuum-molten on it and Heated to 700°C Cut Shows Bond Strength.

FIGURE 4

Isotropic Boron-Nitride Aluminum Pressure sealed to Alumino-Silicate Glass (Corning 1723). Boron-Nitride broken away, shows Fracture within the Material.



a)

b)

FIGURE 3

Graphite(a) and Boron-Nitride(b) Particle Bombardment-Metallized with Aluminum and Brazed with Aluminum

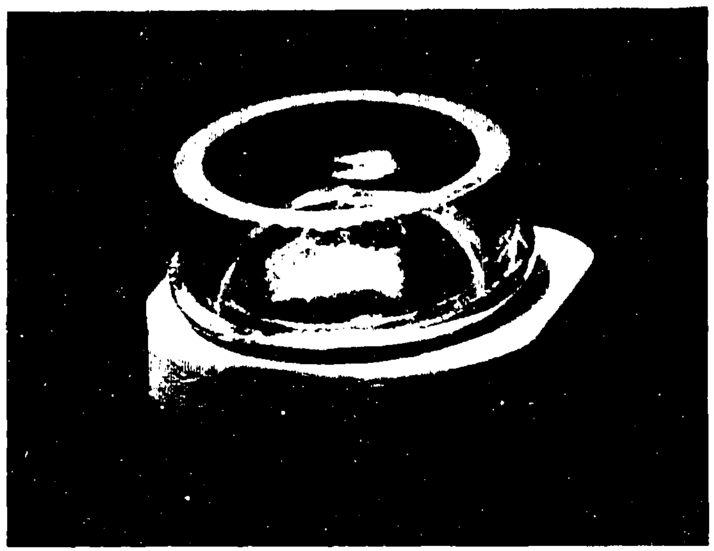


FIGURE 5

Isotropic CVD Boron-Nitride and Alumino-Silicate Glass(Corning 1723) Vacuum-Tight Pressure Sealed with Aluminum Foil.

FIGURE 6

Anisotropic CVD Boron-Nitride Vac.-Tight Pressure Sealed with Aluminum and Torne apart, Showing Bond Strength.

FIGURE 7

Alumino-Silicate Glass(Corning 1723) Vac-tight Pressure Sealed with Aluminum and Broken to Show the Bond Strength.

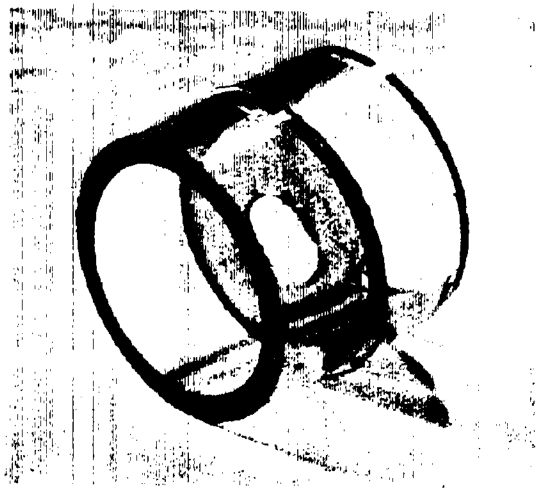


FIGURE 8

Alumino-Silicate Glass(Corning 1723) Vacuum-tight Pressure Sealed with Copper

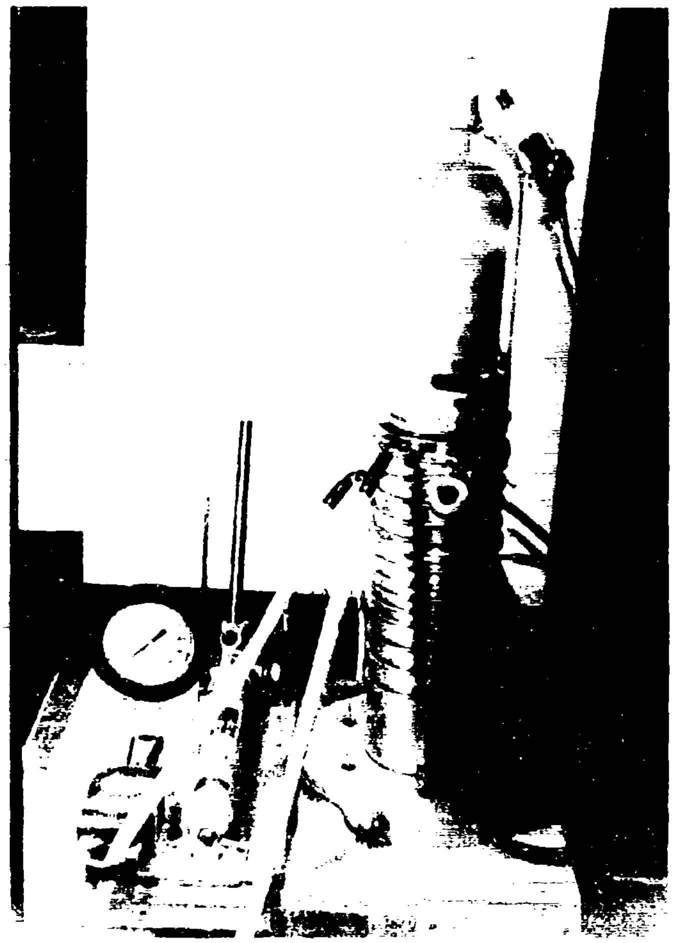


FIGURE 9

Hydraulic Pressing in High-Vacuum and at High Temperatures

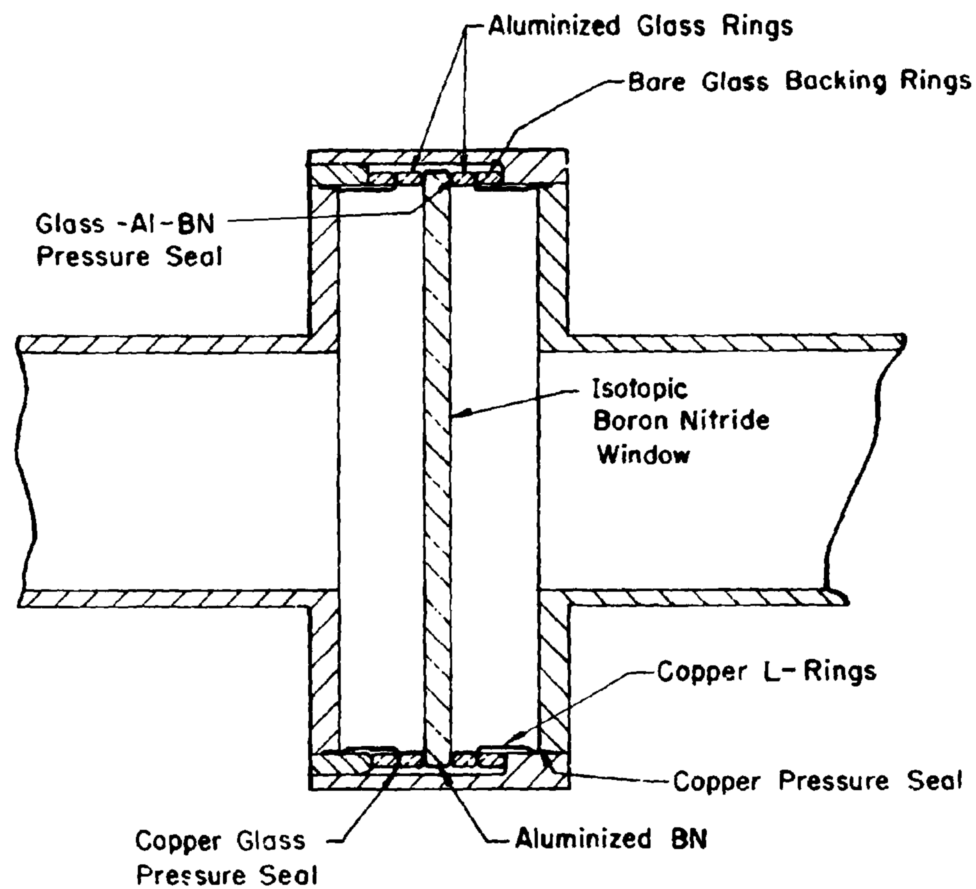


FIGURE 10

Boron - Nitride Window Chamber

Frequency 2850 MHz

Scale 1:1

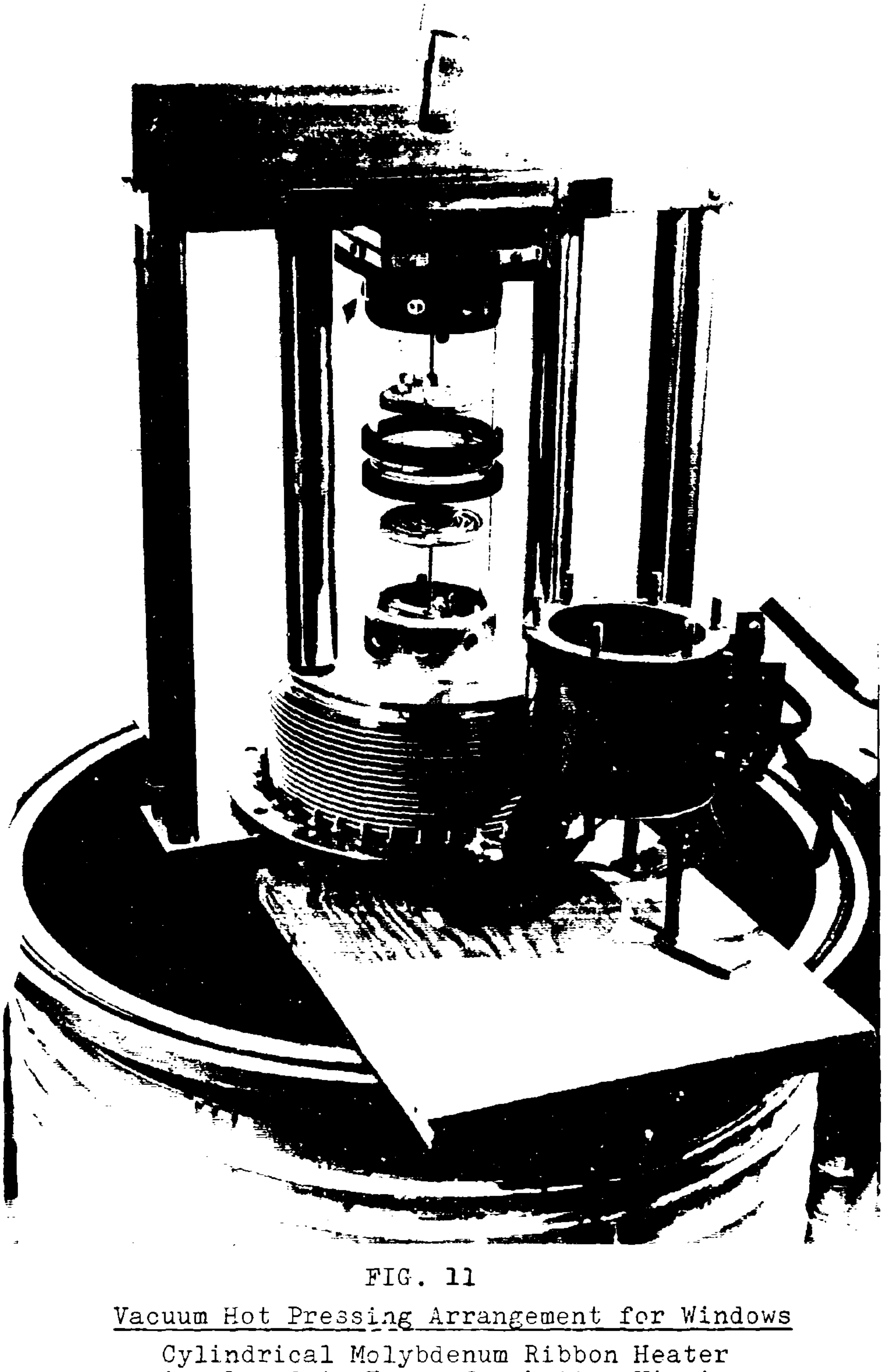


FIG. 11

Vacuum Hot Pressing Arrangement for Windows

Cylindrical Molybdenum Ribbon Heater is placed in Front for better Viewing.

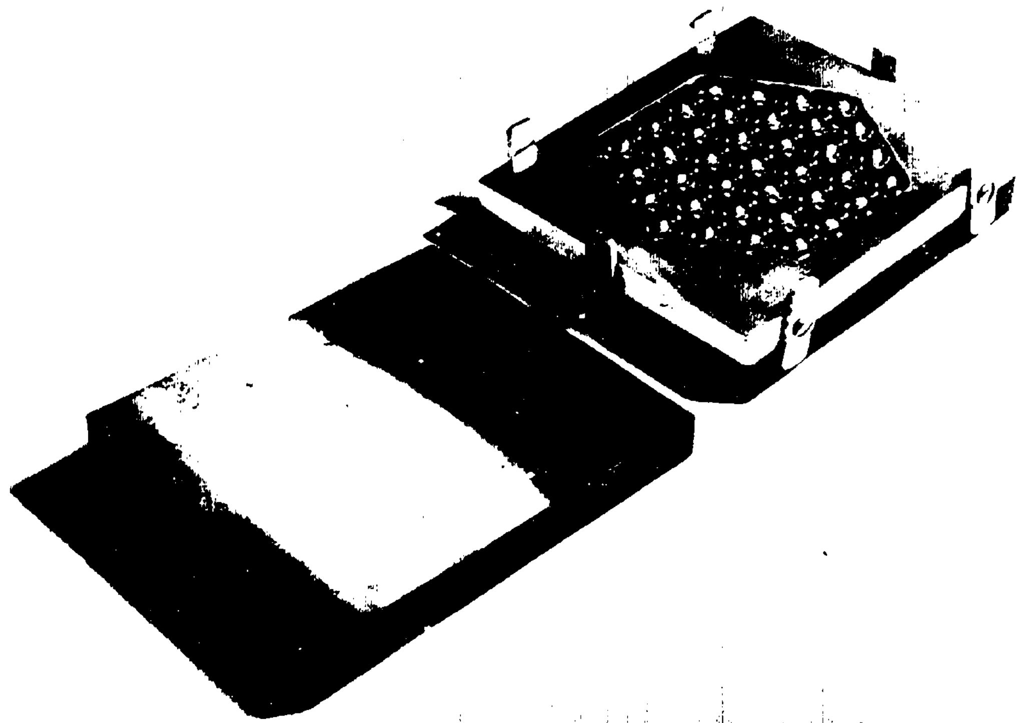


FIG. 12

Pressure Equalizing Pad with Ball Bearing

(allows free lateral motion of pressure tube)

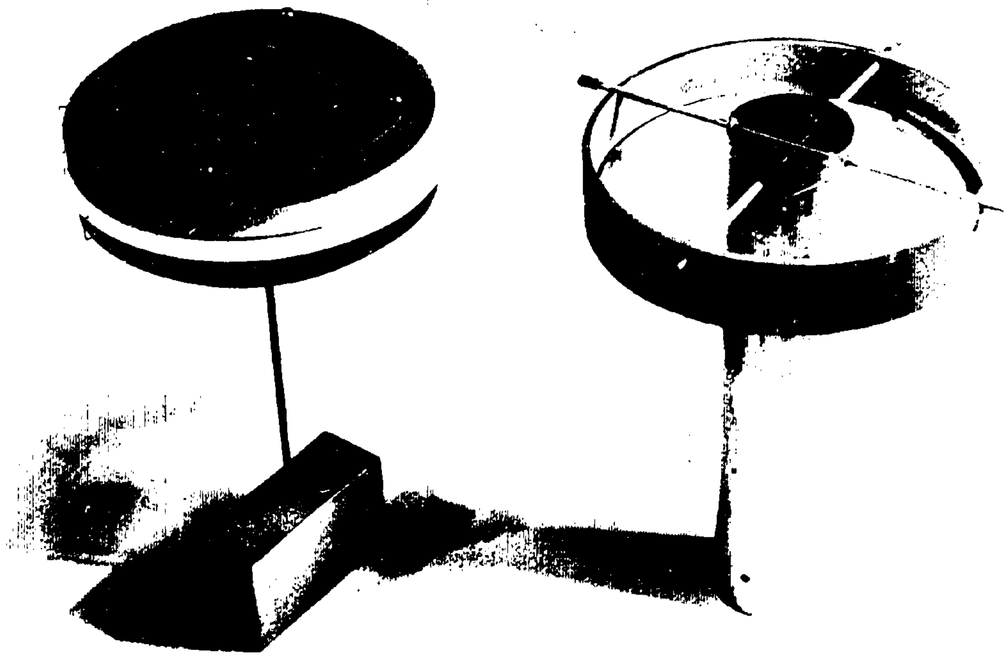


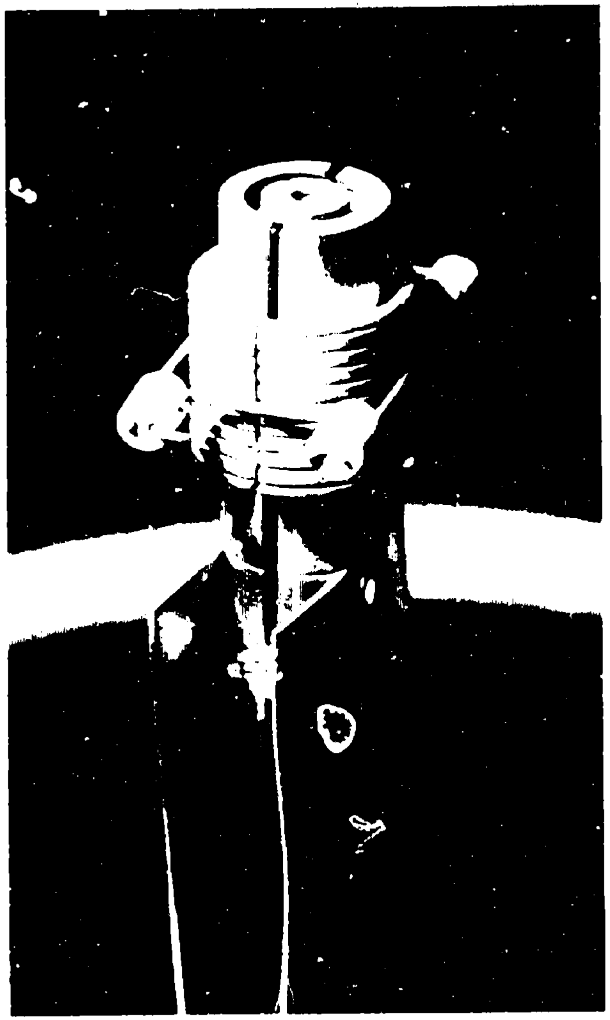
FIG. 13

Graphite Heating and Masking Arrangements for Aluminum Vapor Coating of Window and Backing Rings.

FIG. 14

Pressure Sealed Pyrex Glass Window Assembly.

with Aluminized Pyrex Rings, L-shaped Copper Rings backed by bare Pyrex Glass Rings. At right: Cross-section through Window Seal Area, intentionally broken to show Bonding Strength. Fracture occured within Window Glass and not in Aluminum Seal.



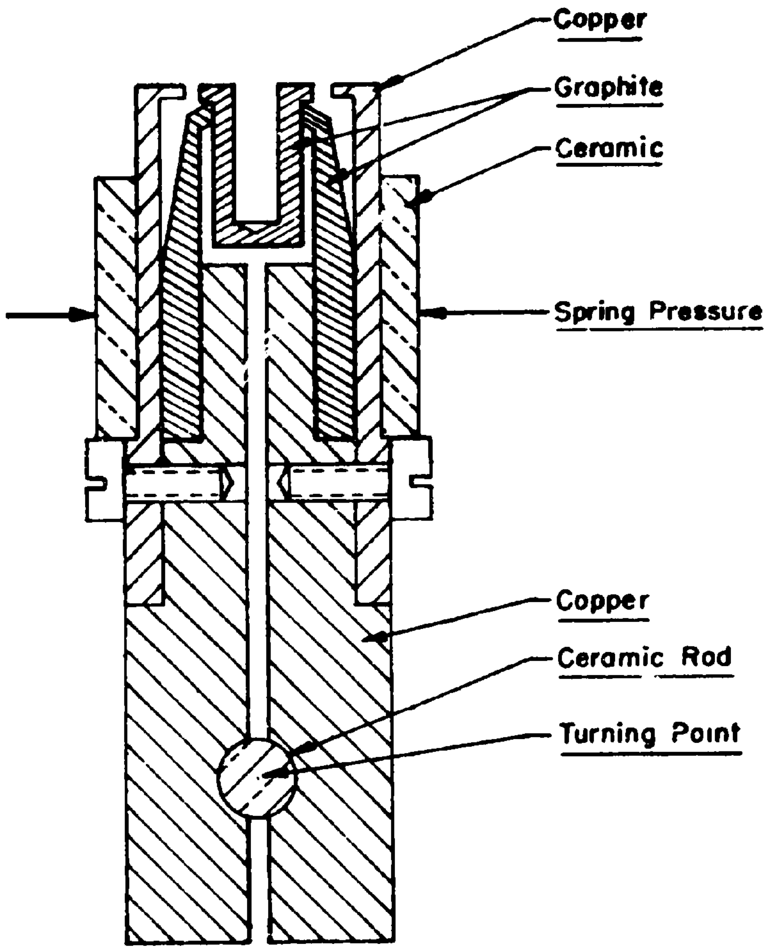


Fig. 15

Graphite Aluminum Vaporizer, Photograph and Cross-Section
Heating by Carbon-Carbon Transition Resistance.

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13. ABSTRACT

The objectives of this program are the application of the metallizing and bonding techniques developed by Heil Scientific Labs under USAECOM contract DA 28-043 AMC-00429(E)[1] to the fabrication and evaluation of high power S-band microwave window assemblies using chemical vapor deposited isotropic boron nitride (CVD-BN) as the window dielectric. Four S-band isotropic CVD-BN poker-chip microwave windows have been furnished by the Techniques Branch, Electron Tubes Division, USAECOM to the Heil Scientific Labs. for this program.

Isotropic boron nitride is the best dielectric for high power microwave windows because of its low dielectric constant, low dielectric losses, high dielectric strength, good heat conductivity and lack of multipactoring. It, however, is difficult to metalize on account of its low thermal expansion, its relative low mechanical strength and its different chemical behavior as a nitride compared with the usual window materials, which are oxides.

Aluminum shows excellent wetting of boron nitride at 1100°C in vacuum. BN can be brazed to itself and to graph[illegible] at this temperature.

After particle bombardme[illegible] with aluminum these brazes can be made at 700°C with good adherance. V[illegible] sure seals to aluminum made at 600°C with or without prior metalizing are st[illegible]d vacuum tight.

Aluminosilicate glasses like Corning 1720 can also be vacuum pressure sealed to aluminum and copper at 600°C on account of their high softening temperature. These glasses match the thermal expansion of isotropic BN. Aluminum pressure seals have bee[n] made to boron nitride with aluminosilicate glass as backing.

Continued on sheet attached

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14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Dielectric						
Microwave Window						
Chemical Vapor Deposited						
Isotropic Boron Nitride						
Particle Bombardment Bonding						
Aluminosilicate glass						
Aluminum						
Copper						
Vacuum-Pressure Seal						
Window Chamber						
Vapor Deposition						
Sealing						

Security Classification

Using the vacuum pressure technique, vacuum tight windows can be assembled using a metallic transition from aluminum to copper, which is backed by glass for vacuum tightness. The glass in these seals is outside of the high frequency field. The current is conducted by aluminum and copper with practically no interface at the metal-boronnitride joint.